

PARTIALLY IONIZED GASES
A REVIEW OF TRANSPORT AND THERMODYNAMIC PROPERTIES*

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ABSTRACT

The thermodynamic properties of dissociated and partially ionized gas in the temperature range greater than about 15,000°K are not generally available in the literature. These properties have now been computed (300 < T < 35,000°K) and comparisons made where possible between the presented data and the data currently available in the literature. The necessary transport property equations which may be utilized in the predictions of these properties are discussed.

INTRODUCTION

The transport processes considered in this work are applicable to any system and it is assumed that one does not know "a priori" the variable composition of the system. In that light, it is necessary to know both the transport properties and the thermodynamic properties of each of the pure species of the system. Thus, this work represents approaches which hopefully will lead to equations appropriate for such evaluations. We consider first the thermodynamic properties of dissociated and partially ionized air.

Thermodynamics

The thermodynamic properties of a gas are of prime importance in heat transfer evaluations and in related hypersonic research work. It is therefore necessary to have generalized expressions which can be programmed (IBM 7090) and used for such computations regardless of the state of the gas (neutral, singly or multi-ionized). The calculations in the report have been performed using such generalized expressions deduced from equations applicable to specific types of species.⁽¹⁾

The total partitions functions, based on quantum statistical theory of heat and statistical thermodynamics, used may be written as:

$$Q_i = \left(\frac{2\pi M_i kT}{h^2 N} \right)^{3/2} \frac{C_i}{V} e^{-\epsilon_i/T} \sum_j f_i g_i e^{-\epsilon_j/T}$$

(Equation 1)

In the above equation, C_1 represents a characteristic quantity, expressed in terms of temperature, for the specie under investigation

$$C_i (\text{atom, neutral}) = \frac{-D_{(\text{molecule})} hc}{2k} = {}^\circ K$$

$$C_i (\text{neutral, } A_2) = 0$$

$$C_i (\text{ATOM, ionized}) = -(I_{(\text{atom})} + \frac{D_{(\text{molecule})}}{2}) \frac{hc}{k}$$

$$C_i (\text{molecule, } A_2^+) = - \frac{(I \times hc)}{2k}$$

$$C_i (\text{Molecule, } AB^+) = \frac{\left(-I_{AB} - \frac{D_A}{2} - \frac{D_B}{2} + D_{AB} \right)}{k} \cdot hc$$

$$C_i (\text{Molecule}) AB = \frac{\left(\frac{D_A}{2} - \frac{D_B}{2} + D_{AB} \right) hc}{K = k} \cdot h.c.$$

Equation I was differentiated twice with respect to $\frac{1}{T}$ to yield the following two equations:

$$Q_i' = \left(C_i - \frac{3}{2} T \right) Q_i - \left[\frac{2\pi M_i kT}{h^2 N} \right]^{3/2} \frac{C_i}{v_e} \sum_j \epsilon_j f_j e^{\epsilon_j/T}$$

$$Q_i'' = \left(C_i - \frac{3}{2} T \right) \left[Q_i' - \left(\frac{2\pi M_i kT}{h^2 N} \right)^{3/2} \frac{C_i}{v_e} \right]$$

$$+ \sum_j \epsilon_j f_j g_j e^{\epsilon_j/T} + 3/2 T^2 Q_i$$

$$+ \left(\frac{2\pi M_i kT}{h^2 N} \right)^{3/2} \frac{C_i}{v_e} \sum_j \epsilon_j^2 f_j g_j e^{\epsilon_j/T}$$

(Equation III)

The specific heats were calculated using the expression given below: (*)

$$C_V = \frac{R}{T^2} \left[\frac{Q''}{Q} - \left(\frac{Q'}{Q} \right)^2 \right] \quad (\text{Equation IV})$$

The energy values used in these computations were obtained from various references which represent in our opinion the best available data (2) (3) (4) (5). In comparing the data computed from the equations above, with published data, one finds alarming disagreement between the data. The disagreements are due to many reasons - namely (a), the number of energy levels included in the evaluation of the partition functions, (b), the reliability of these levels and (c), whether or not the coupling effects are introduced. The omission of these effects which become appreciable at high temperatures can cause serious errors in the evaluation of these properties⁽⁶⁾. The parameter f_j in equation I corrects for these effects and it may be expressed as:⁽¹⁾

$$f_j = \frac{1}{\sigma_j \left(1 - e^{-u_j} \right)} \left[1 + \frac{\sigma_j}{3} + \frac{\delta_j \gamma_j}{\sigma_j \gamma} + \frac{\delta}{e^{-\mu_j - 1}} + \frac{2 \chi_e \mu_j}{\left(e^{-u_j} - 1 \right)^2} \right]$$

where:

$$\sigma_j = \frac{B_e h c}{k T} \left(1 - \frac{1}{2} \delta_j \right); \quad u_j = ch \omega_e (1 - 2 \chi_e)$$

$$\delta_j = 6 \frac{B_e}{\omega_e} \left[\left(\frac{\omega_e \chi_e}{B_e} \right)^{1/2} - 1 \right]$$

(Equation V)

In the evaluation of the partition function of the atoms where there are a multiplicity of states, the choice of the "cut-off" is very important. The results computed here include all energy levels which made any significant contribution to the partition function.

* Thermodynamics for Chemists - Glasstone's P. 102 - 3rd Edition -
D. Van Nostrand & Co., Inc., 1948.

Figures I, II and III show the molecular specific heats in comparison with available data; figures IV, V and VI, comparison data for the molecular ions; figures VII and VIII, comparison data for the neutral atomic species and figures IX and X show the comparison data for the atomic ions.

It will be observed that in the case of NO and O₂, divergence in the data begins at relatively low temperatures. In the case of oxygen, close agreement is seen to exist up to about 13,000°K. Beyond that temperature Beckett's data does not continue to increase at the same rate. Browne's data follows Beckett's but is lower at 10,000°K where he discontinued computations. Hansen's data is not expected to follow either of the other two curves because his computations did not include the excited states. As a result, his data reaches the maximum and levels off at about 7 cal/mole°K. In the temperature range > 10,000°K, Beckett's data is lower than the reported data. This is probably due to the difference in the two approaches. Beckett utilized a Morse function to determine the intermolecular forces for the diatomic constituents in the evaluation of the ideal gas properties in the range 5,000 to 25,000. In addition, he used the second virial coefficients for his evaluations in the range from 5,000 to 100,000°K. Glasstone's approach which was appropriately modified to include the coupling effects, was used in this work. All known molecular states were included in the evaluation of the partition functions. The two approaches are not compatible at high temperatures. According to Glasstone (*) the technique employed in this work is adequate for the computation of the heat capacities at any temperature. Another essential quantity necessary in hypersonic investigations is the internal energy function. This function may be obtained directly from the partition functions or by an integration of the specific heats. These data are shown in figures XI and XII.

Transport Properties

The state of the art of transport property calculations for neutral molecular interactions is in a satisfactory condition. Many papers have been published (7) (8) (9) (10) (11) (12) on the transport properties of molecular gases (non-polar) and one can say that there is reasonable agreement among

them. It is recognized, however, that potentials like the L. J. and some of the approximate ones cannot be expected to yield values of a high degree of accuracy at extremely high temperatures. However, improvements are continuously being made both in theoretical approaches as well as in experimental techniques. More and more experimental and theoretical data is appearing in the literature. Some of these data, although performed at relatively low temperatures ($< 3000^{\circ}\text{K}$) present significant advancements in the over-all art. From these data, one can estimate appropriate cross-sections and evaluate a true potential energy function $\phi(r)$.^{(13) (14)} Some serious work of a theoretical nature has been in the investigation of the interaction potentials between two ground state oxygen atoms. Dalgarno⁽¹⁵⁾ has derived explicit expressions for the thermal conductivity and viscosity of atomic oxygen. At elevated temperatures, he points out that the transport properties are controlled by the short range repulsive forces and that the spin-orbit interaction energy is small compared to the energy of thermal motion and to the electrostatic interaction energy. Analytic expressions for the potential energy curves at small separations corresponding to the eighteen known spectroscopic states have been given by Mason, Hirschfelder and Vanderslice (a, b, c). For temperatures between 100°K and 2000°K , Dalgarno estimates the thermal conductivity to be $67.1 \times 10^{-7} \text{ erg}/^{\circ}\text{K cm sec}$ and the viscosity to be accurately represented as $3.34 \times 10^{-7} \text{ micropoise}$. Unfortunately, the estimation of the properties charged-charged and charged-neutral particles is much less developed and there are many uncertainties in the interaction potentials. In many cases, only approximate potential forms are available. The types of potential which have the greatest success in predicting the transport properties of plasma are listed in figure XIII. Some attempts in treating a complex system

-
- a. Vanderslice, J. T., Mason, E. A., and Maisch, W. C., J. Chem. Phys. 33, 614 (1960).
 - b. Vanderslice et al - J. Chem. Phys. 32, 515 (1960).
 - c. Kinowlow, D., Hirschfelder, J. - Phys. of Fluids 4 637 (1961).

of this type have been successful and thus represent significant contributions to this area. (16) (17) (18) (19) (20).

The interaction between charged particles, being coulombic in nature, presents special difficulties. These difficulties are due to the long range nature of the forces which cause divergence in the cross-section integrals. The technique of avoiding this difficulty is to introduce some type of cut-off. This has been done by many authors (16) (18) (21).

The potential energy function for the attractive shielded coulombic potential is:

$$\begin{aligned} Q(r) &= -\frac{c}{r} e^{-n/h} & r > \sigma \\ &= \infty & r < \sigma \end{aligned} \quad (\text{Equation VI})$$

For purposes of simplifying the computation, it is convenient to use dimensionless parameters wherever possible.

The above equation can be rewritten as:

$$Q(r) = -\frac{\epsilon}{r^*} e^{-a(r^* - 1)}$$

where:

$$r^* = \frac{r}{\sigma}; a = \frac{\sigma}{h}; \epsilon = \frac{ce^{-\sigma/h}}{\sigma}$$

$$h = \sqrt{4\pi N_j e_j^2 / k T}$$

Another important parameter in the evaluation of the cross-sections is the transport integral, $\Omega(l, s)$, the evaluation of which depends upon the angle of deflection x . If one makes the substitution $b^* = \frac{b}{\sigma}$, one can write the standard H. C. B. (Hirschfelder, Curtiss, Bird) expression for the angle of deflections as:

$$x = \pi - 2b^* \int_{r_m^*}^{\infty} \frac{dr^*}{r^{*2} \left[1 + \frac{2\epsilon}{u g^2 r^*} e^{-a(r^*-1)} - \frac{b^{*2}}{r^{*2}} \right]^{1/2}}$$

(Equation VII)

r_m^* the distance of closest approach becomes the lower limit of integrations. The solution of the denominator of the integrand, when set to 0, may be accomplished by numerical technique. A combination of the Newton's and the method of False position is recommended.

If one makes the substitution $-g^{*2} = \frac{\mu g^2}{2\epsilon}$, it is seen that the angle of deflection becomes a function of the variables $-b^*$, g^* and \underline{a} which depend upon the electron density and σ , the collision diameter.

Finally, one can write the expression for the omega integral

$$\frac{2}{(r+1)! T^{*(s+2)}} \int_0^{\infty} e^{-g^{*2}/T^*} g^{*(2s+3)} Q^{(l)*} dg^*$$

(Equation VIII)

where:

$T^* = kT/E$ and

$$Q^{(l)*} = \frac{2}{\left[1 - \frac{1}{2} \frac{1 + (-1)^l}{1+l} \right]} \int_0^{\infty} (1 - \cos^l x) b^* db^*$$

(Equation IX)

It is seen that equation IX yields infinite values for $\Omega^{(l,s)*}$ when \underline{a} approaches 0. This is due to the fact that the potential reduces to that of an unscreened coulombic and as such the cross-sections diverge. Work is in progress on the evaluations of these equations for a large number of pressures and the temperature range up to 25,000°K. Liboff⁽¹⁶⁾ computed the $\Omega^{(l,s)}$ integrals

for the coulombic and the attractive shielded coulombic potential. Similarly, he uses an effective shielding distance - the Debye shielding distance. Others (17) (18) (20) (21) (22) (23) (26) have used cross-sections of the shielded coulombic type to study ionic mobilities as well as to evaluate the transport properties of fully ionized gases. Hence, one may state that the technique of utilizing the Debye shielding distance rests upon rather sound theoretical grounds. Some of the basic work on this phenomena dates back to the investigations of Persico⁽²⁴⁾.

Although the problem of solving the problems of charged-charged interaction is difficult, problems of an even greater magnitude are faced in studying interactions involving charged-neutral and neutral-neutral interactions. Some of the work done by Hassé⁽²⁶⁾, Hassé and Cook⁽²⁵⁾, Meador⁽²⁷⁾, Amdur and Mason⁽¹¹⁾, Hirschfelder (HCB) ⁽²⁸⁾, Mason et al may be considered as a starting point in such investigation.

The collision integrals of Hasse and Hasse and Cook may be used to investigate forces of the form - $\frac{d\phi}{dr} = \frac{\mu}{r^5}$ or the corresponding potential - $\phi(r) = -\frac{c}{r^4}$.

The standard HCB⁽²⁸⁾ $\Omega^{(2,2)*}$ integrals may be obtained from tabulated relationships from Hassé's work⁽²⁵⁾. Thus, for a charged induced dipole type of function - $\frac{1}{r^4}$, the $\Omega^{(2,2)*} = 4 \frac{X}{\lambda}$. The parameter λ is $\sqrt{T^*}$ which depends upon the potential energy function and X which is a function of the cross-section, velocity, and the angle of deflection X. Once λ is determined then one can obtain the corresponding value for X. Integrals corresponding to $\Omega^{(1,1)*}$, $\Omega^{(1,2)*}$ and $\Omega^{(1,3)*}$ can be estimated from the work of Mason and Schamp⁽¹⁷⁾ from a knowledge of $\sqrt{T^*}$. Integrals for repulsive atom-atom interactions can be estimated from a combination of the work of Monchick⁽²⁹⁾, Bade, Mason & Yun⁽³⁰⁾ and Peng et al⁽³¹⁾.

The Omega integrals and the dimensionless quantities A*, B* and C* for the repulsive potential N-N are shown below.

$T^{\circ}\text{K}$	$\sigma^2 \Omega(2, 2)$	$\sigma^2 \Omega(1, 1)^*$	$\sigma^2 \Omega(1, 2)^*$	$\sigma^2 \Omega(1, 3)$	A*	B*	C*
300	12.11	10.26	9.499	8.946	1.180	1.141	.9255
1,050	9.107	7.590	6.930	6.458	1.200	1.161	.9173
5,050	5.935	4.80	4.287	3.918	1.235	1.199	.9818
10,000					1.253	1.122	.8785
24,800					1.282	1.256	.8556

As shown by Peng⁽³¹⁾, the A^*_{ij} and B^*_{ij} may be considered as constants. Our calculations show that to be true. It is to be noted that only a repulsive potential is apparently important in this interaction. The calculations above were made utilizing only the ${}^7\Sigma_{\mu}^+$ state of the nitrogen atom.

Acknowledgments

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NOMENCLATURE

$$A_{ij}^* = \frac{\Omega^{(2,2)*}}{\Omega^{(1,1)*}}$$

$$B_{ij}^* = \frac{\left[5 \Omega^{(1,2)*} - 4 \Omega^{(1,3)*} \right]}{\Omega^{(1,1)*}}$$

$$C_{ij}^* = \frac{\Omega^{(1,2)*}}{\Omega^{(1,1)*}}$$

g_j = Statistical weight

D_e = Dissociation energy

I_e = Ionization energy

N_e = Electron density

h_D = Debye Shielding distance = $\left[kT / 8 \pi N_e \epsilon^2 \right]^{1/2}$

χ_e = Anharmonicity constant

ω_e = Equilibrium vibrational frequency of the particle

B_e = Rotational constant

M = Molecular weight

r_m = Distance of closest approach

ϵ_j = Energy of the j^{th} state expressed in degrees $^{\circ}\text{K}$. (Electronic)

h = Planck Constant = 6.624×10^{-27} erg. sec

k = Boltzmann constant = 1.38047×10^{-16} erg. deg. $^{-1}$.

α = $B_e \delta$

L.J. = Lennard - Jones 6:12 Potential

HEAT CAPACITY

N₂

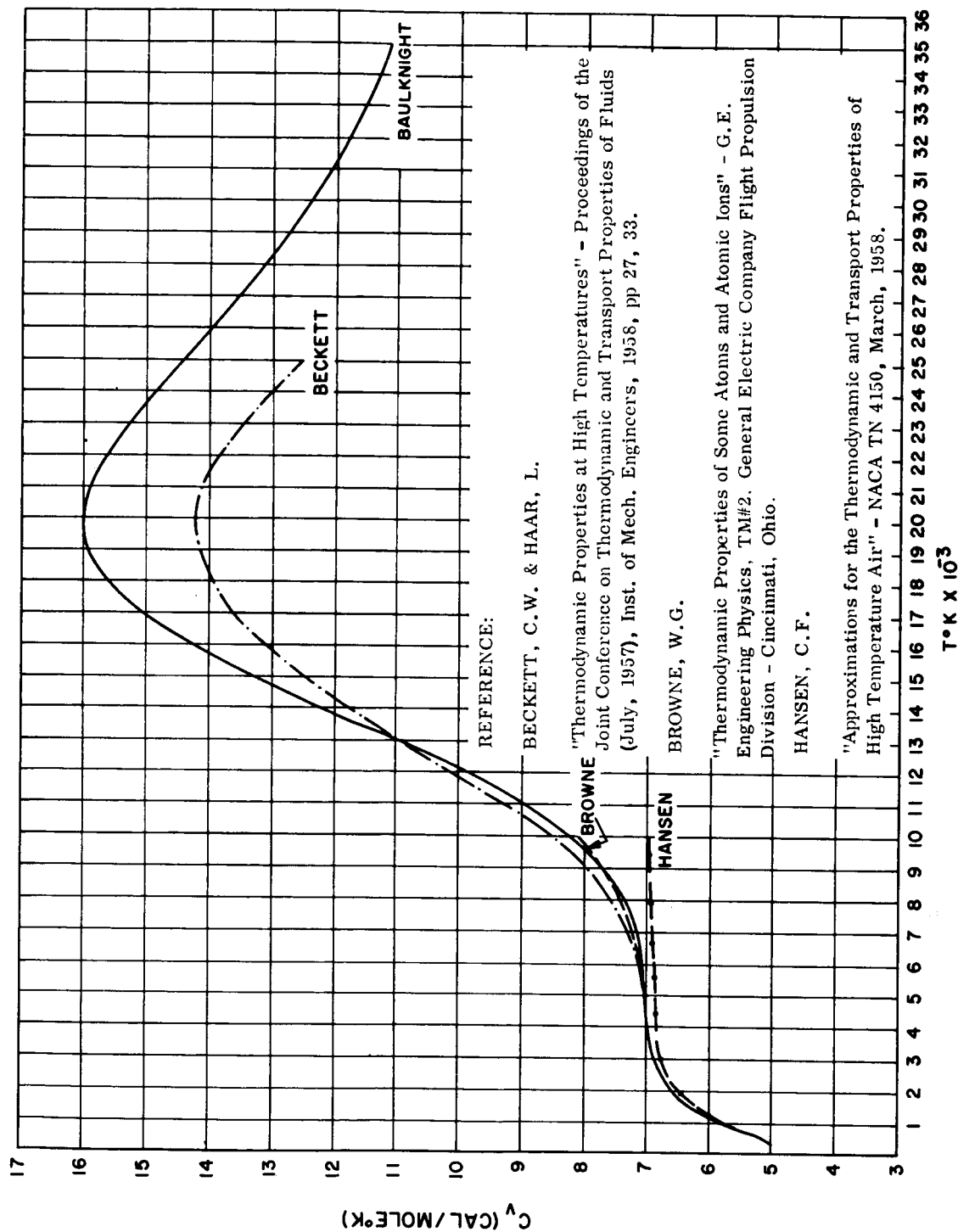


FIGURE I

SPECIFIC HEAT

O₂

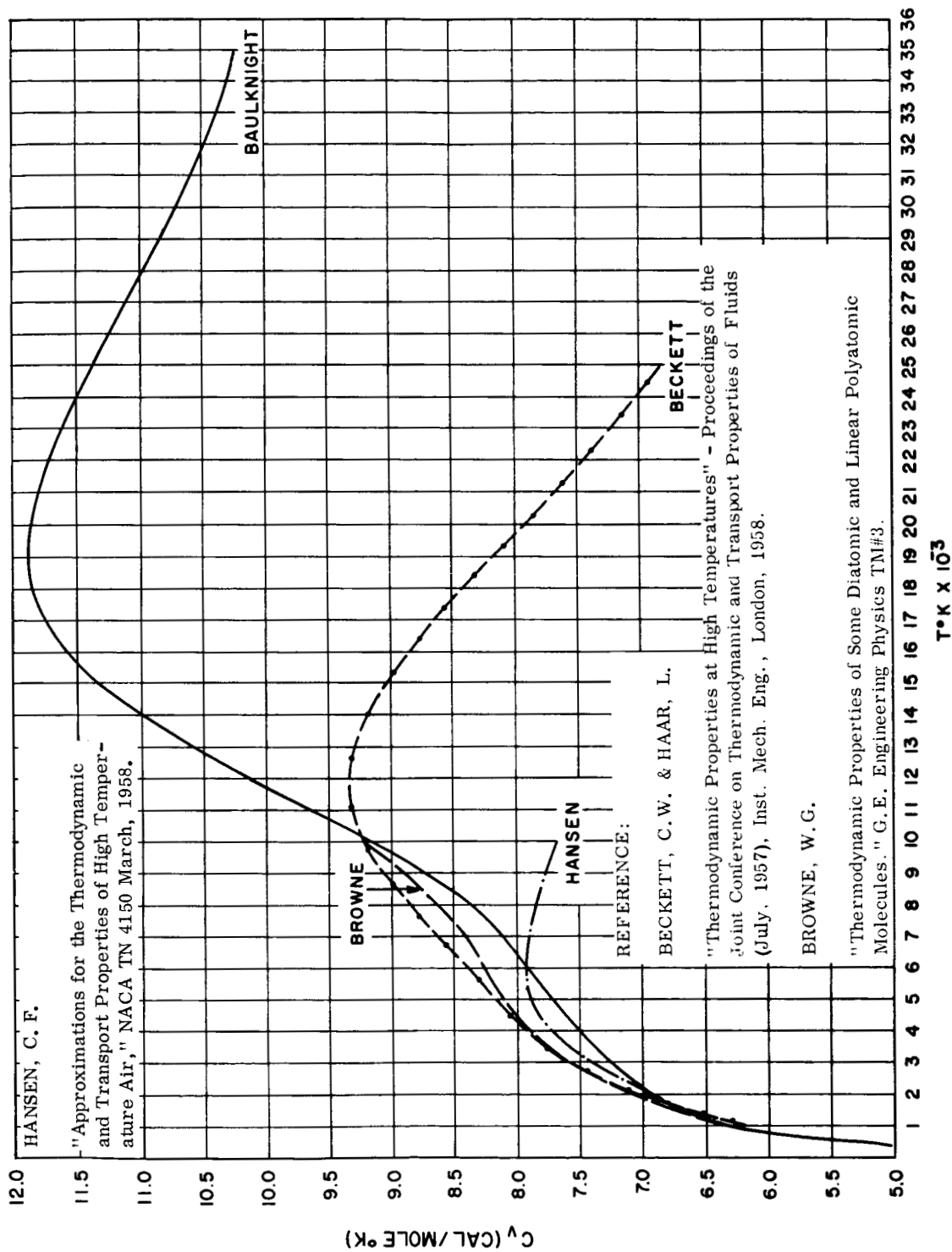
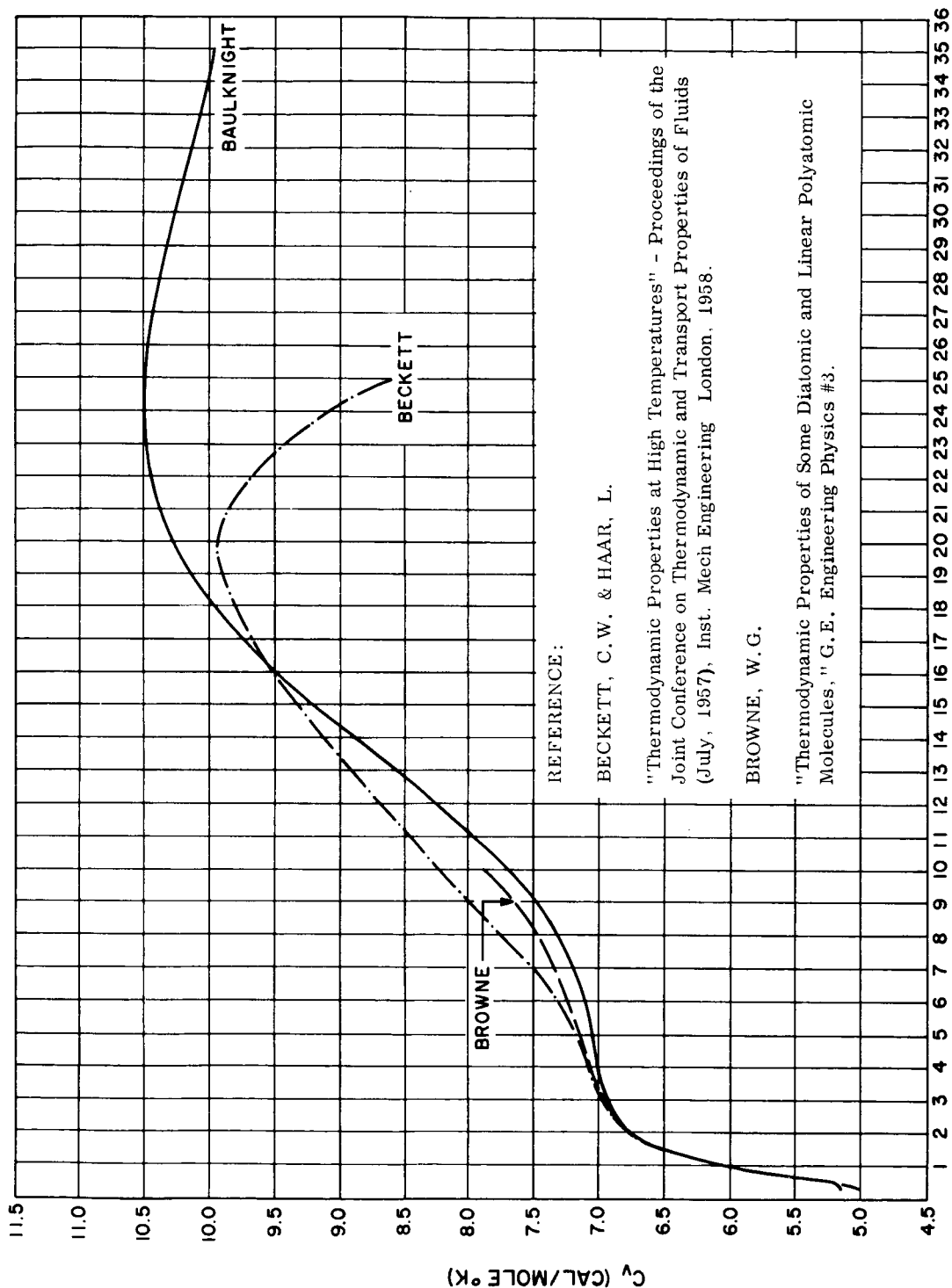


FIGURE II

SPECIFIC HEAT
NO



$T^{\circ}K \times 10^3$

FIGURE III

REFERENCE:

BECKETT, C.W. & HAAR, L.

"Thermodynamic Properties at High Temperatures" - Proceedings of the Joint Conference on Thermodynamic and Transport Properties of Fluids (July, 1957), Inst. Mech Engineering London, 1958.

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"Thermodynamic Properties of Some Diatomic and Linear Polyatomic Molecules," G.E. Engineering Physics #3.

SPECIFIC HEAT
 N_2^+

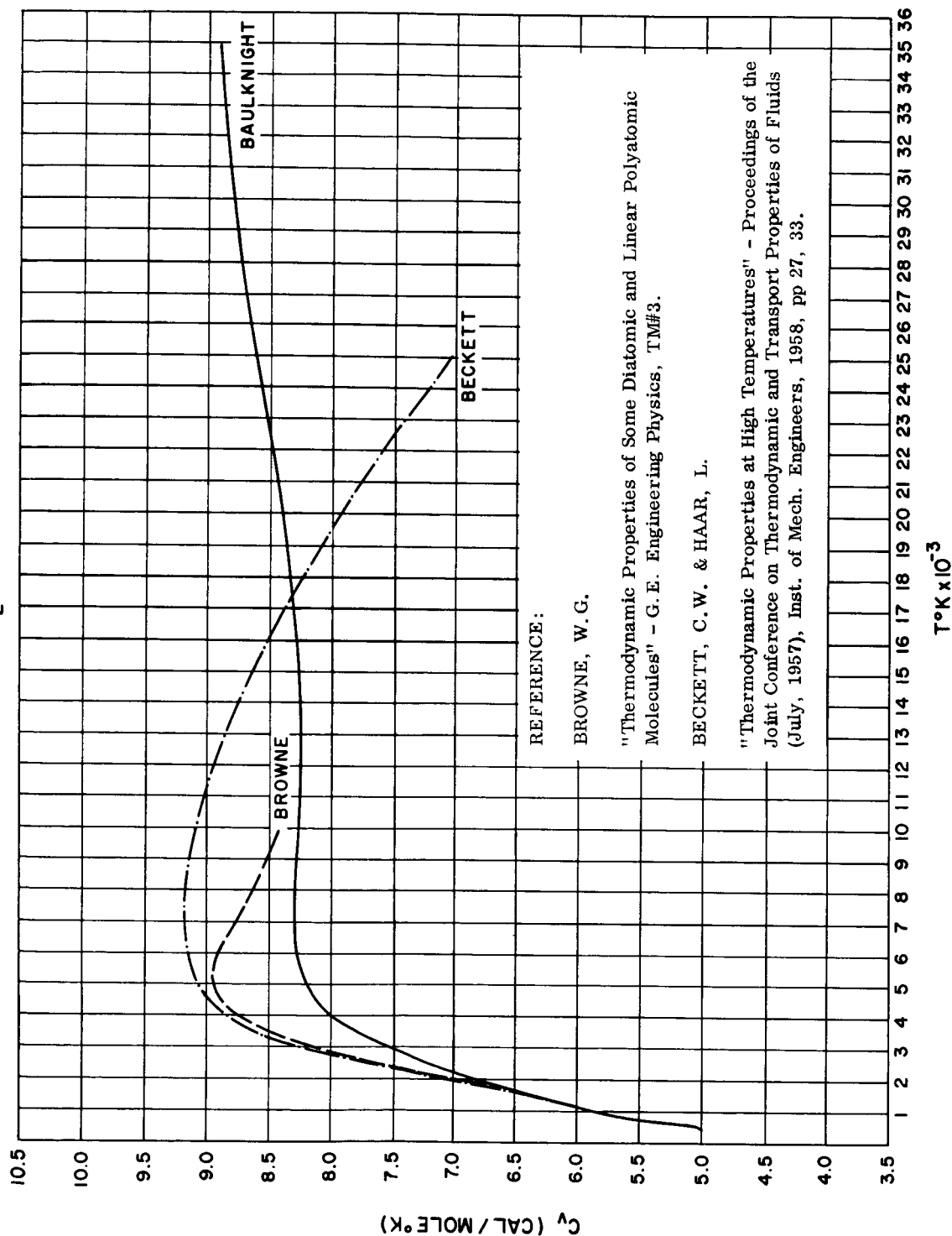
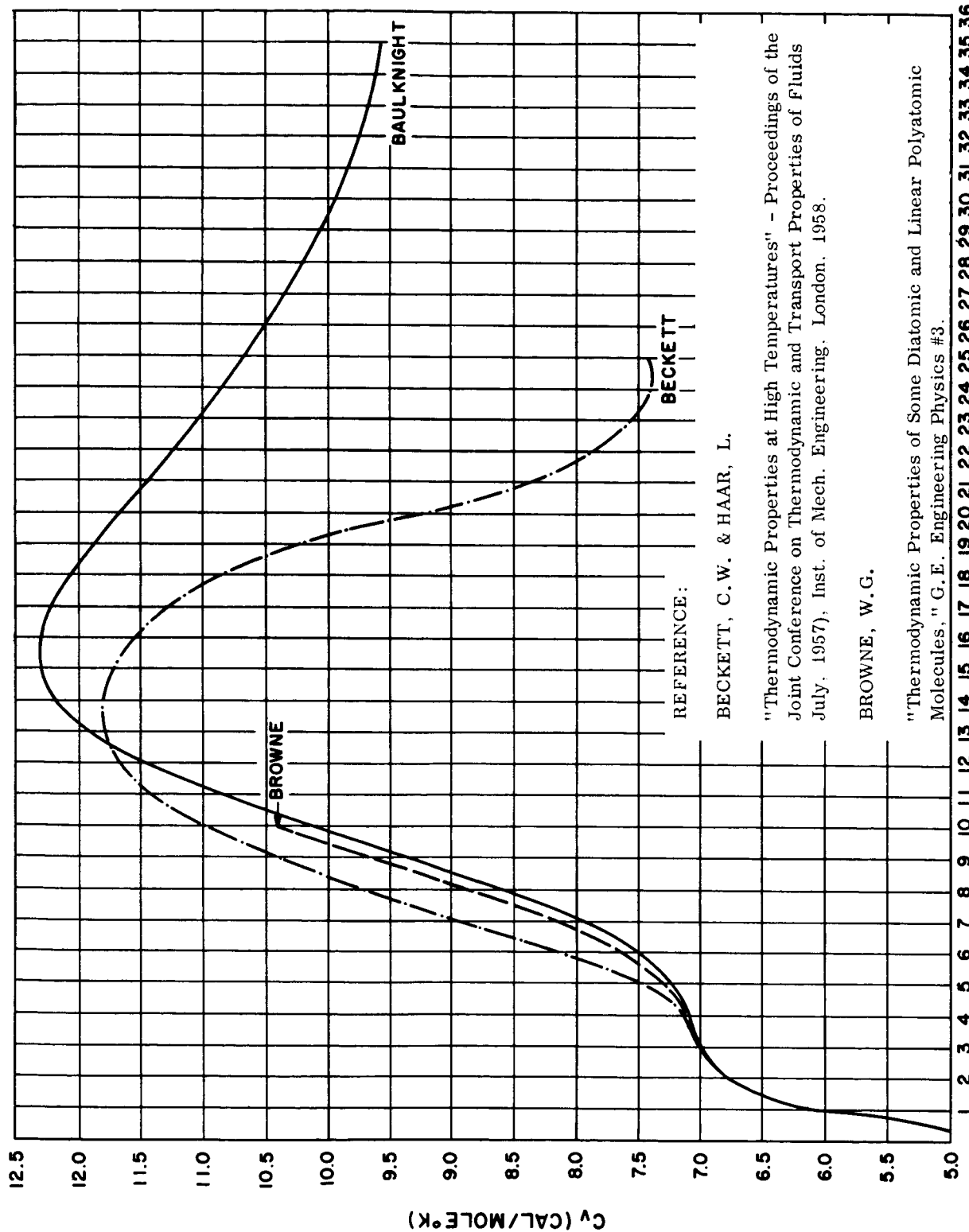


FIGURE IV

SPECIFIC HEAT
 O_2^+



$T^{\circ}K \times 10^3$

FIGURE V

SPECIFIC HEAT
NO⁺

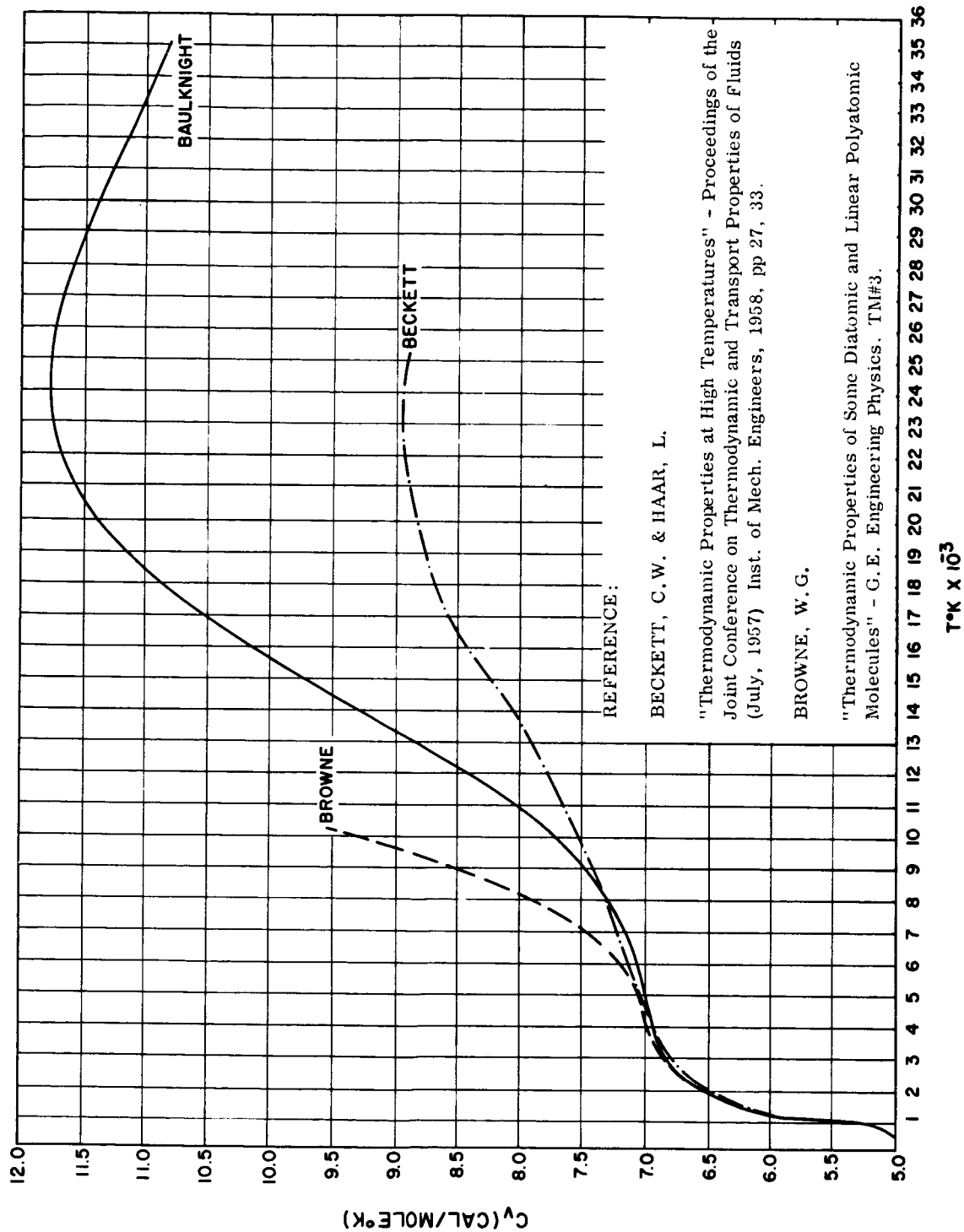
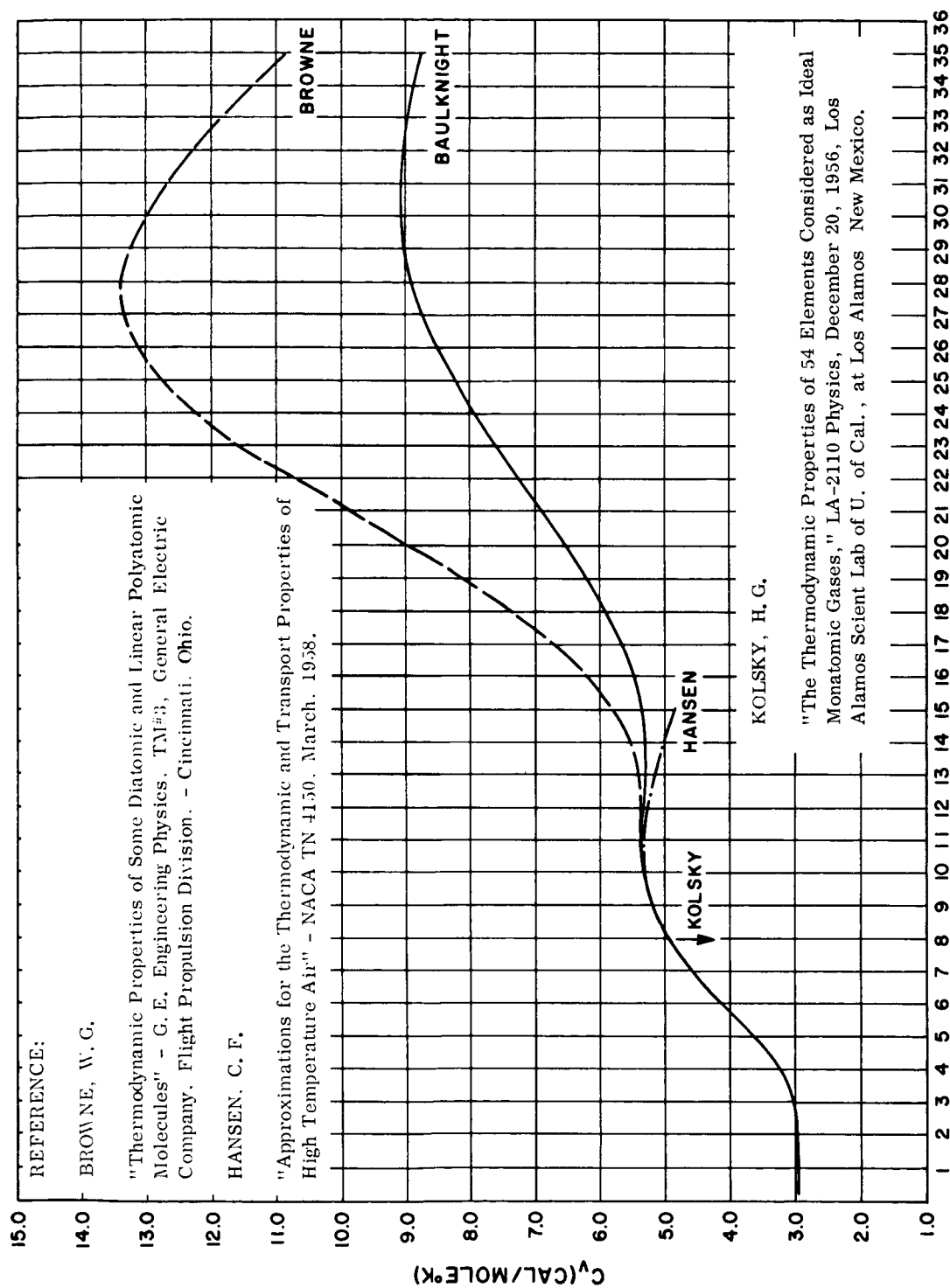


FIGURE VI

SPECIFIC HEAT

N



$T^{\circ}K \times 10^{-3}$

FIGURE VII

SPECIFIC HEAT

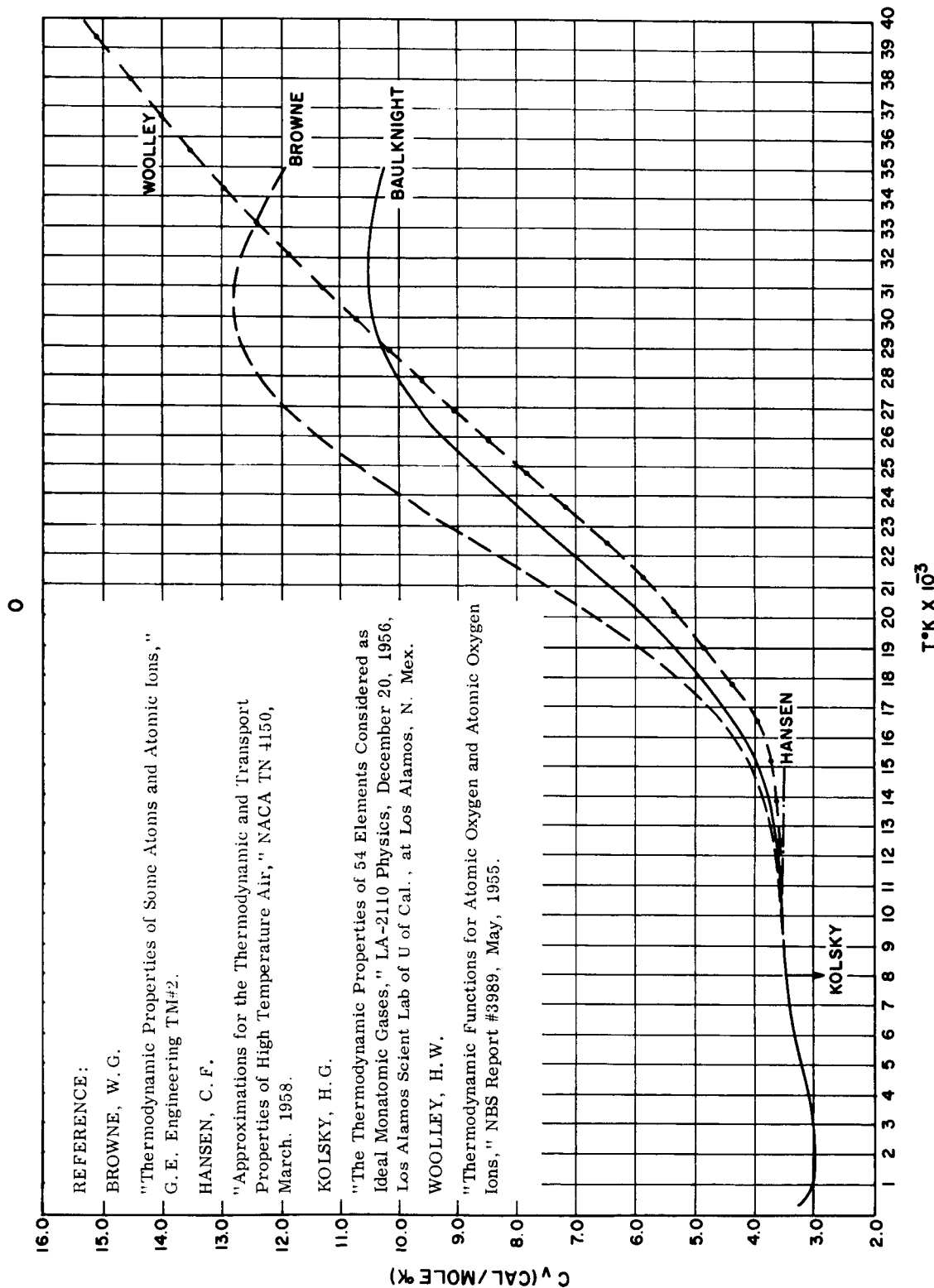


FIGURE VIII

SPECIFIC HEAT

N^+

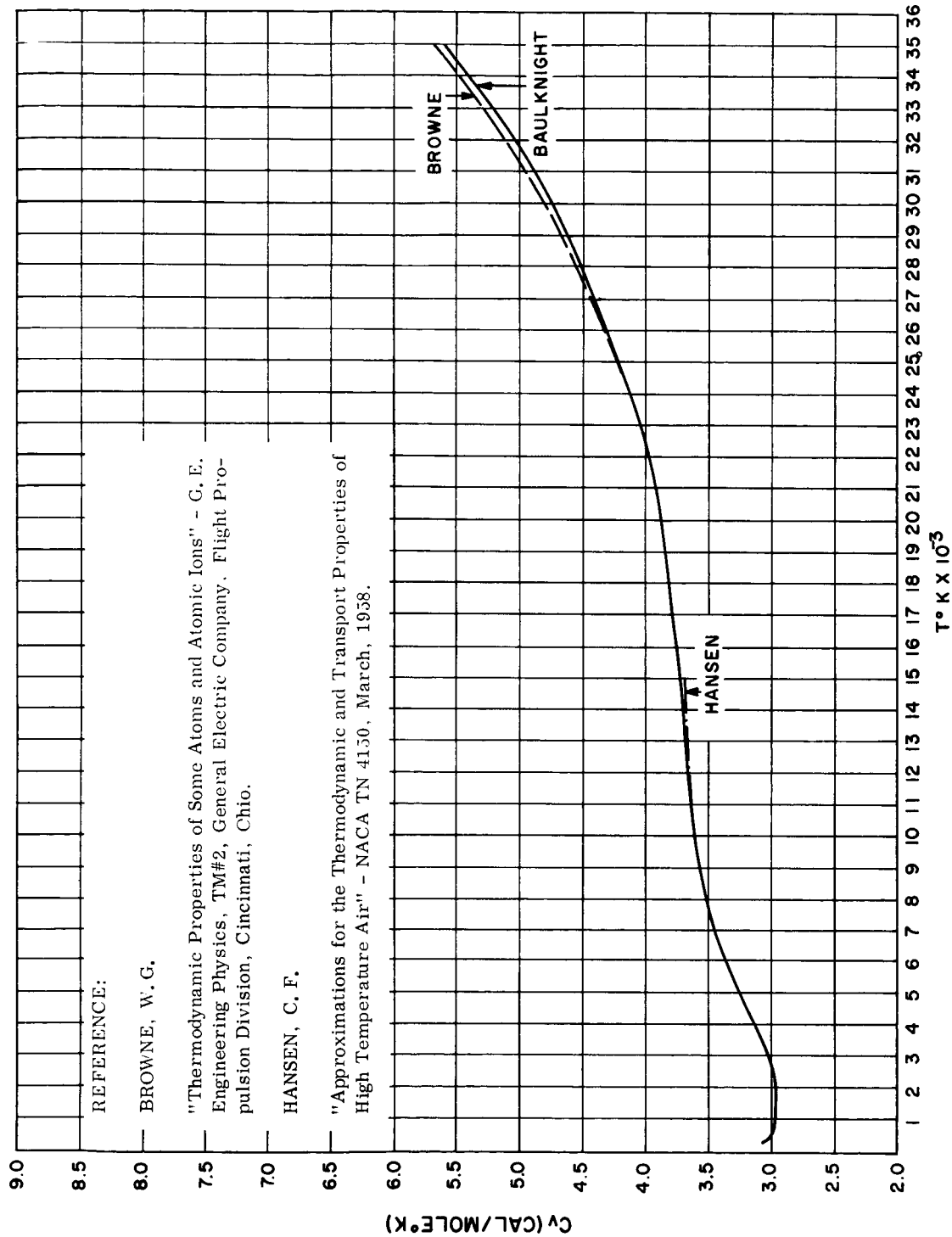
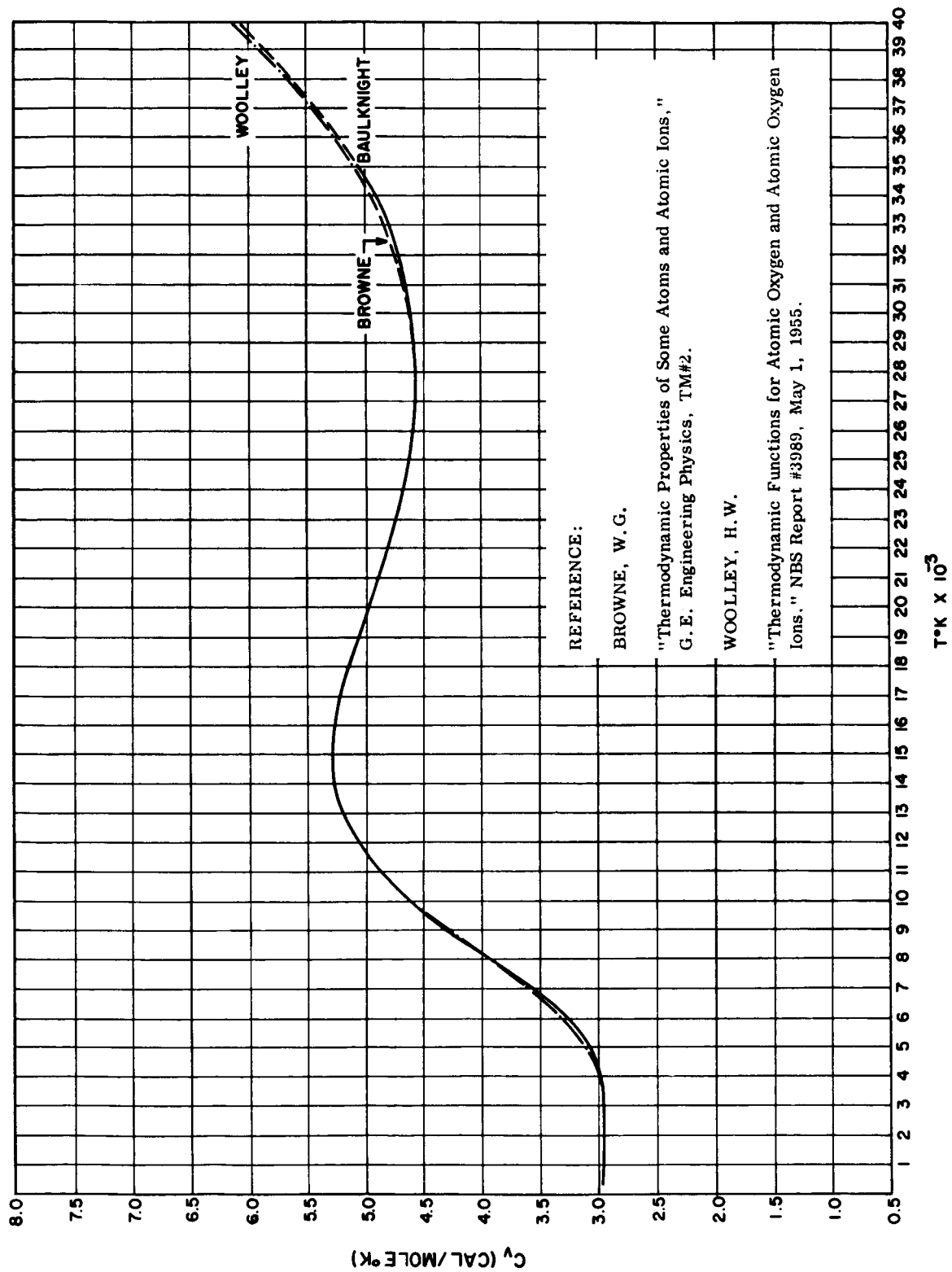


FIGURE IX

SPECIFIC HEAT O⁺



REFERENCE:

BROWNE, W. G.

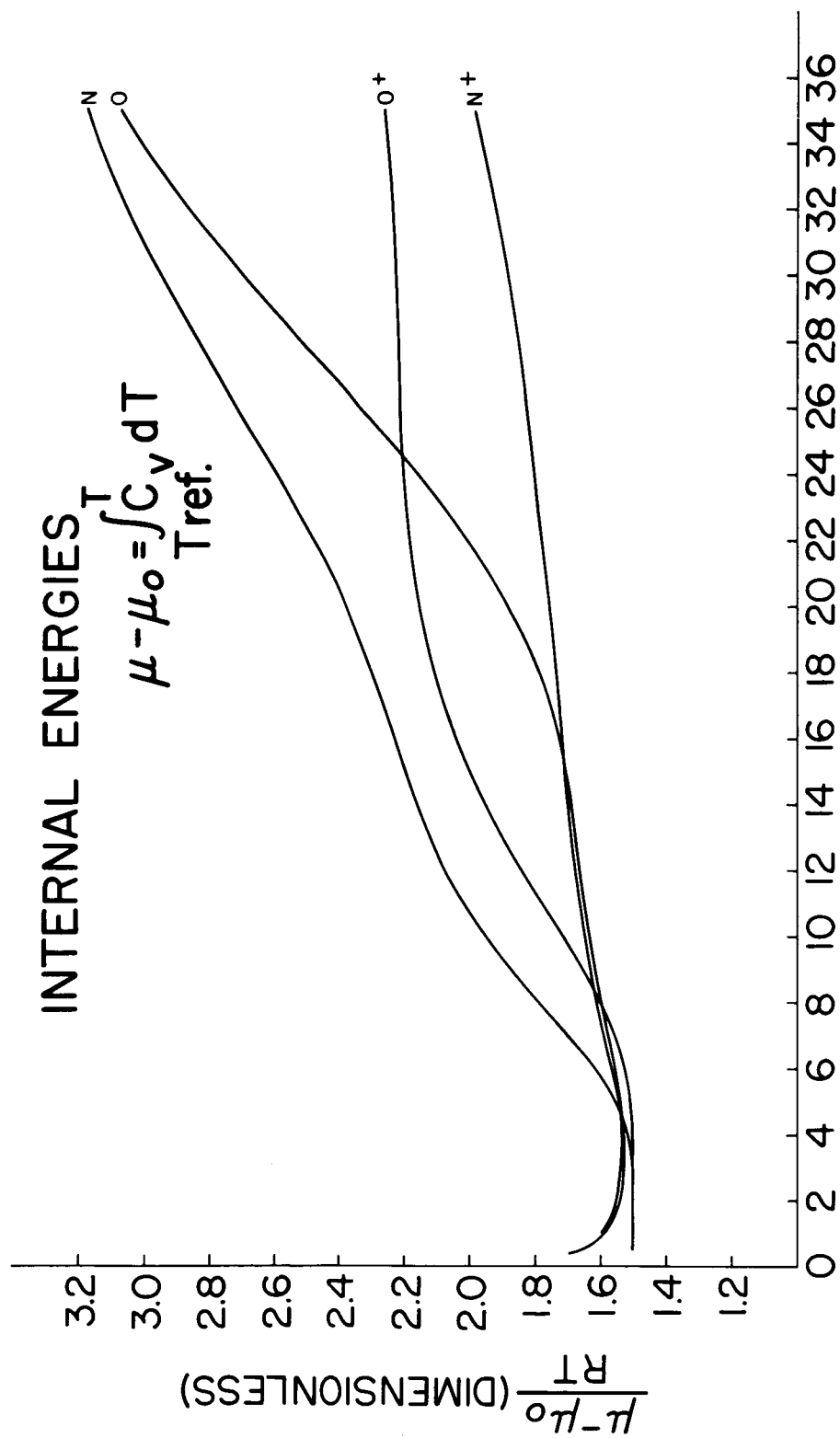
"Thermodynamic Properties of Some Atoms and Atomic Ions,"
G. E. Engineering Physics, TM#2.

WOOLLEY, H. W.

"Thermodynamic Functions for Atomic Oxygen and Atomic Oxygen
Ions," NBS Report #3989, May 1, 1955.

$T^\circ K \times 10^3$

FIGURE I



$T \text{ } ^\circ K \times 10^{-3}$

FIGURE XI

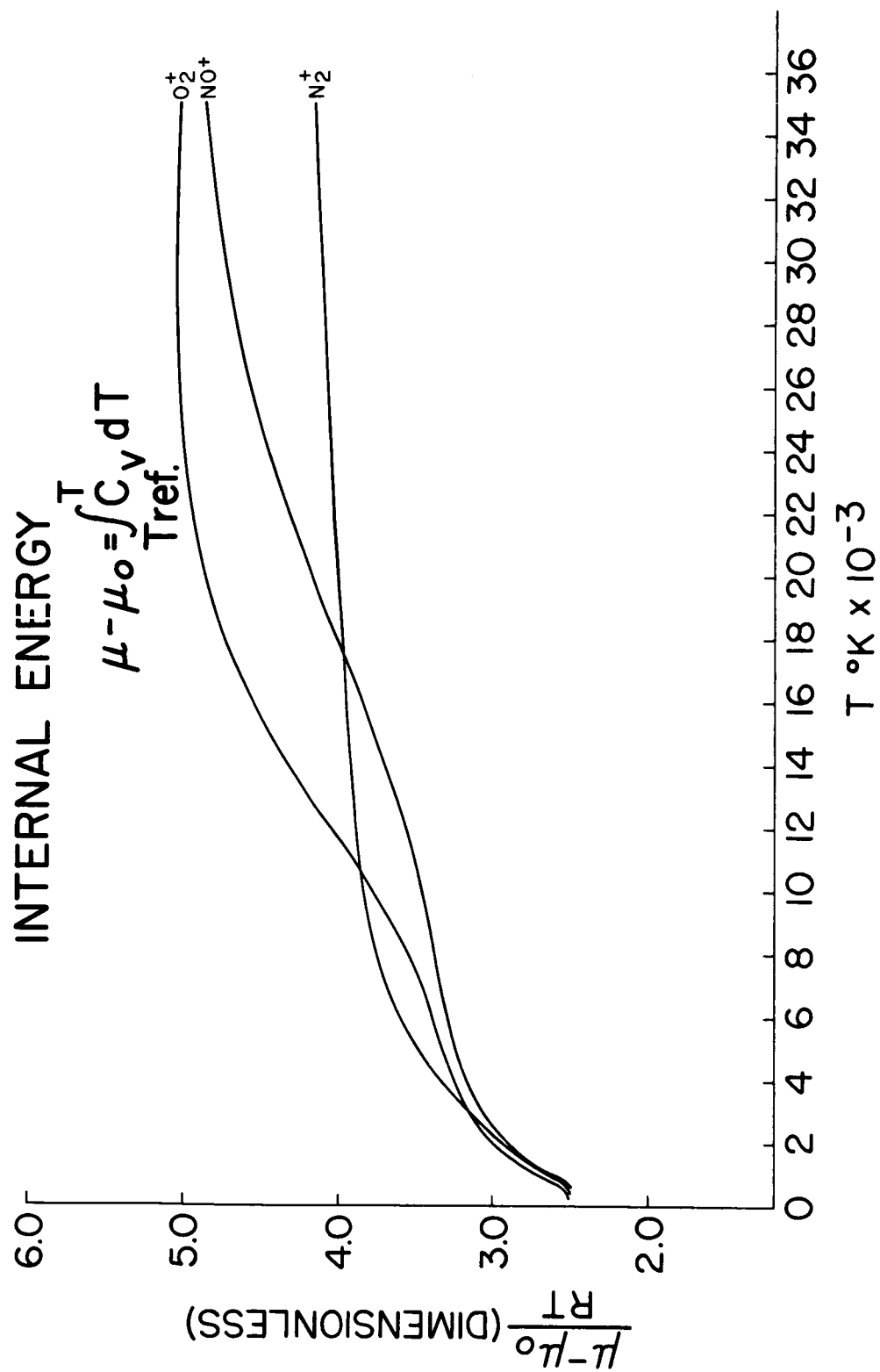


FIGURE XII

INTERMOLECULAR POTENTIAL HIGH TEMPERATURE TRANSPORT PROPERTIES

(ENCOUNTER)

STATE	PARTICLES	TYPE	APPROPRIATE POTENTIAL ϕ_r
NEUTRAL	MOLECULES AND ATOMS	SELF	BUCKINGHAM EXP. 6 $\phi(r) = \frac{\epsilon}{1 - \frac{\epsilon}{\alpha}} \left[\frac{6}{\alpha} \exp\left(\alpha \left[1 - \frac{r}{r_m}\right]\right) - \left(\frac{r_m}{r}\right)^6 \right] - \left(\frac{r_m}{r}\right)^6$ $= \omega(r \leq r_{max})$
CHARGED	A^+, M^+, \bar{e}	SELF	SHIELDED COULOMBIC REPULSION $\phi(r) = AC^{-r/h} \quad r > \sigma$ $= \omega \quad r > \sigma$ (h = DEBYE LENGTH)
NEUTRAL	$\left\{ \begin{array}{l} M+M \\ M+A \\ A+A \end{array} \right\}$	BINARY	BUCKINGHAM EXP. 6 { COMBINATION OF THE ATTRACTIVE PORTION OF THE BUCKINGHAM AND A REPULSIVE POTENTIAL OF THE FORM $\phi(r) = Ae^{-r/r^*}$
CHARGED	$\left\{ \begin{array}{l} M+A^+ \\ M+M^+ \\ M+\bar{e} \\ A+A^+ \\ A+M^+ \end{array} \right\}$	"	REPULSIVE POTENTIAL OF THE FORM ABOVE.
	$\left\{ \begin{array}{l} M+\bar{e} \\ A+A^+ \\ A+M^+ \end{array} \right\}$	"	CHARGE INDUCED DIPOLE $-\phi(r) = \frac{\epsilon^2 \alpha}{2r^4}$
	$\left\{ \begin{array}{l} A^+M^+ \\ A^+\bar{e} \\ M^+\bar{e} \end{array} \right\}$	"	SHIELDED COULOMBIC REPULSION - $\phi(r) = \epsilon_1 \epsilon_2 / r \quad \exp(-r/h)$
		"	"
		"	ATTRACTION -
		"	"

FIGURE XIII

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